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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6 : B01J 19/00, 13/02, G03F 7/00, G09F 3/00

A1

(11) International Publication Number: WO 99/47254

(43) International Publication Date: 23 September 1999 (23.09.99)

(21) International Application Number: PCT/GB99/00762

(22) International Filing Date: 15 March 1999 (15.03.99)

(30) Priority Data:

9805508.0	13 March 1998 (13.03.98)	GB
9805510.6	13 March 1998 (13.03.98)	GB
9805512.2	13 March 1998 (13.03.98)	GB

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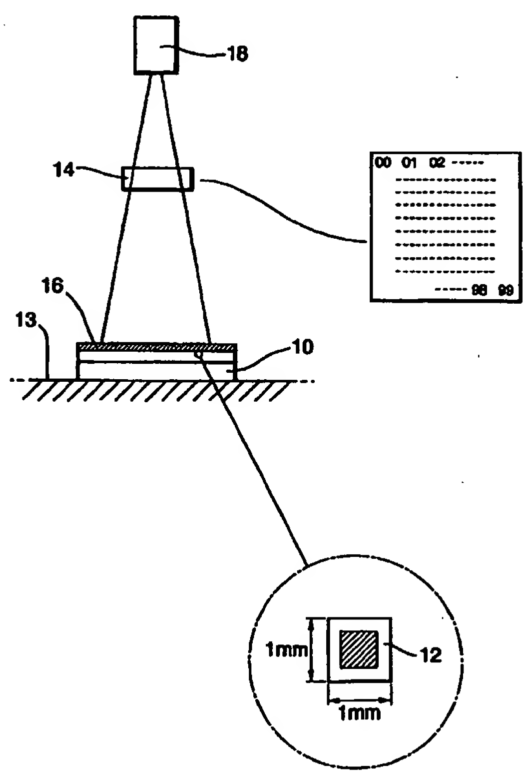
(81) Designated States: CA, JP, KR, SG, US, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

Published
With international search report.

(54) Title: APPARATUS FOR, AND METHOD OF, MANUFACTURING A PLURALITY OF UNIQUELY LABELLED ARTICLES

(57) Abstract

Apparatus for and a method of manufacturing a large number (typically more than 10^6) articles (12) are described. Typically 10,000 articles (12) are defined on a substrate (10) in an array of 100 rows by 100 columns. One method uses a screen printing technique and a mask to form a unique indicia on each article of the substrate. Several substrates (10) may be formed like this. Second, possibly third and more indicia is then applied to all articles (12) on a substrate (10). A different indicia is applied to all articles (12) on a different substrate (10). The result is that a very high number of unique articles (12) are fabricated rapidly and relatively easily. An alternative method employs a photolithographic technique. The two different methods may be combined. The substrates (10) are diced or cut into uniquely labelled articles (12) and may be used in combinational chemistry. The invention overcomes problems associated with prior art systems in that a large number of labelled articles (12) can be fabricated using less steps.



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Apparatus for, and m thod of, manufacturing a plurality f uniquely labelled articles

The present invention relates to an apparatus for, and method of, manufacturing a plurality of uniquely labelled articles. More particularly but not exclusively, the articles are smaller than 10 mm x 10 mm x 10 mm, and preferably smaller than 2 mm x 2 mm x 2 mm. Said uniquely labelled, articles (or beads as they are sometimes called) are capable of being used in the area of chemistry known as combinatorial chemistry.

Combinatorial chemistry is a technique whereby very many different chemical compounds are produced by multiple chemical reactions. It is desirable in combinatorial chemistry to label uniquely each chemical compound. Until recently this has been a very difficult task. Typically additional, sophisticated chemical processing stages had to be undertaken to add a "chemical tag" to each bead. Such chemical tags have proved difficult to identify and, moreover, have restricted the type of chemical processes which may be employed in formation of desired compounds.

Recently the use of a bead consisting of a composite solid phase support and a physical tag has been developed. An example is disclosed in UK Patent Application GB-A-2 306 484 (Tracey). However, the aforementioned UK Patent Application does not disclose all the steps required to produce the beads and is therefore considered to be non-enabling.

Published International Patent Application WO-A2-9712680 (Irori) describes a combination matrix having a recording means which is smaller than about 10 mm square and which supports an optically readable code. Methods of marking code are described.

Other prior art methods, although successful, had limitations, particularly with regard to the time required to fabricate the beads. Advances in combinatorial chemistry increasingly require labelling of hundreds of millions of compounds. Allowing for unique labelling of many synthesis sets leads to a requirement to select from as many as 10^{12} unique labels or codes (hereinafter "indicia"). Increasingly this requires combinatorial chemistry tagging methods to be more sophisticated.

It is an object of the present invention to provide an apparatus for, and method of, manufacturing a plurality of uniquely labelled articles. The articles may be beads, for example, for use in combinatorial chemistry.

According to the present invention there is provided a method of fabricating a plurality of articles, each having a unique label, comprising the steps of: defining the articles on at least two substrates; forming or causing to be formed on articles, of each substrate, a first indicia sufficient to identify articles one from another on each substrate; forming a second indicia on articles on one of said substrates sufficient to identify articles of one substrate from articles of another substrate; and disassembling the substrates into a plurality of labelled articles.

Preferably articles are formed in rows and columns and thereby define an array. An example is an array of 100 rows by 100 columns, giving 10000 individual beads or articles. If ten such arrays are prepared and each article in an array is given a second indicia, then 1000,000 different beads will have been produced from only 11 different sets of means for applying indicia and two process steps applied to each set of articles.

According to a another aspect of the present invention there is provided a method of manufacturing a plurality of uniquely labelled articles comprising the steps of: applying a medium, via a first printing screen to a substrate; removing the first printing screen presenting the substrate to a second printing screen and applying a medium via said second printing screen to said substrate; curing said substrate and cutting the substrate into a plurality of uniquely labelled articles.

Preferably the number of articles or beads produced exceeds thousands and most preferably millions. Preferably the medium applied via the or each printing screen is a paste and is of suitable viscosity to maintain a defined shape of the aperture through which it is urged. Bonding of the medium may be by way of drying, heating, firing, or curing or a conversion method for example using ultra violet (UV) or another form of radiation. to cure the medium.

If defects lead to unreadable encodings then this is of no consequence as such articles or beads may be selectively discarded before chemical processing begins.

Uniquely labelled articles, are preferably smaller than 10 mm x 10 mm, preferably as small 250 μm x 250 μm. Currently a size in the range 500–1000 μm along a side is most preferred.

According to a further aspect of the present invention there is provided a method of manufacturing a plurality of uniquely labelled articles, comprising the steps of: exposing a photoresist coated substrate, from which the articles are to be formed, using a first photomask to define a first set of unique low order indicia at first locations on the substrate; subsequently exposing the substrate to a second photomask to define high order indicia at second locations on the substrate, so that combinations of low order indicia and high order indicia are unique for each article and disassembling the substrate into said plurality of labelled articles.

Labelled articles may be formed using for example, screen printing then different codes may be formed using one or more photomasks. The advantage of this combined process is that a large number of articles or beads are formed and each article can be arranged to carry a sufficient amount of compound to enable combinational chemical reactions to occur.

Preferably a third photomask is used in conjunction with said first (low order) photomask and said second (high order) photomask. The third photomask is termed a middle order photomask and provides yet further unique indicia to the article.

Most preferably, first and second middle order photomasks are used. In this case the high order and low order photomasks are combined to form a single mask.

Photomasks are preferably of the type used in photolithography. In addition, further middle order photomasks may be used. Typically middle order photomasks comprise indicia, which may be, for example, numerals 0-9, located at different positions thereon. Indicia may be located at an orientation corresponding to a lower, left hand corner of a surface on each article or bead. A set of 10 photomasks, each of which bears a single number selected from the range 0-9, may be used to produce an image of the number in any of 100000 bead positions with an array of 100 x 100 rows and columns of articles. Alternatively, a set of 100 photomasks each having a number in the range 00-99 may be used with an array of 100 x 100 rows and columns of articles. The total number of possible combinations is therefore 1,000,000 different beads from a total of 100 photomasks.

In a particularly preferred embodiment, semiconductor wafers on which beads are defined, are sequentially, exposed firstly to a series of low order photomasks. Development of the resist occurs after exposure. Subsequently, wafers are exposed to higher order photomasks. Each photomask pattern is configured so that the area of photoresist coating material to be exposed is in the region of the low order numerals exposed, and no other resist in the bead area is exposed.

Outside the area of substrate used to form beads there may be positioned, alignment features which allow subsequent photomasks to be aligned to the first exposure, thereby achieving registration one with another.

In addition photomasks may be rotatable with respect to a substrate. Thus by judicious choice of geometry of photomask it is possible to further increase the number of combinations of articles which can be produced.

After exposure a reversal bake of the substrate is preferably performed. This baking process may be preferred to resist development because it is simpler and cheaper. Baking renders exposed substrate material insoluble in developer and has minimal effect on unexposed material.

Wafers may be divided into lots after an exposure. All wafers in each of these lots are exposed to one or more middle order photomask(s). The middle order photomasks are aligned to the pre-existing resist pattern using the aforementioned alignment means. After exposure a second reversal bake may be performed.

Wafers are then re-arranged in a combinatorial fashion to form for example 10 lots of 10 wafers each, so that no two wafers from the previous lot (in any previous stage) are in the same lot at a subsequent stage. This type of fabrication means that even more beads can be produced with less masks, but more steps are required.

During exposure of the middle order photomask, if it is aligned so that it is offset slightly from the position previously used, a second middle order photomask is exposed. Again a reversal bake is preferably performed. Following this reversal bake wafers are preferably subjected to a flood exposure which renders soluble all of those areas of resist which

have not been subjected to an exposure followed by a reversal bake. Following the flood exposure the resist is again developed. This second developing stage results in a plurality of articles on each wafer, each article carrying a unique pattern or code. The patterns may be transferred into an underlying layer for example a metal layer by a simple etching process. The resist is then removed. Apparatus for performing the methods is also provided, as specified in claim 19.

Ways in which the invention may be performed will now be described, by way of examples only, and with reference to the Figures in which:

Figure 1 shows a diagrammatic sectional view of a sketch of a screen printing apparatus;

Figures 2 a to e show diagrammatically successive views illustrating a method of fabrication of uniquely labelled articles;

Figure 3 is a diagrammatic view of an alternative apparatus for producing uniquely labelled articles;

Figure 4 is a diagrammatic view of an article, showing an overlay of an active area using a high order photomask for a bead whose label is "103123237514".

Figure 5 is a diagrammatic view of the article in Figure 4, showing an overlay of an active area and a low order photomask;

Figure 6 is a diagrammatic view of the article in Figure 4, showing an overlay of an active area showing a first middle order photomask;

Figure 7 is a diagrammatic view of the article in Figure 4 showing an overlay of an active area and a second middle order photomask; and

Figure 8 is a diagrammatic view of the article of Figure 4 carrying the final label "103123237514".

Referring to Figure 1, there is now described a method for fabricating a set of uniquely labelled articles or beads by a screen printing method.

Tile or substrate 10 defines a substrate from which articles or beads 12 are to be formed. A first printing screen 24 overlays the tile 10. Screen 24 has defined in it a plurality of individually identified gaps or apertures (not shown). Each gap or aperture represents a recognisable code, such as, for example, a decimal digit or group of digits. Location of each screen is by way of a mechanical rest or other well know technique.

Paste 20 is applied to the surface of screen 24 and a squeegee 22 is drawn across the surface of the screen 24 in the direction of arrow A. Paste 20 is sufficiently viscous to remain in a volume defined by the aperture, through which it has passed, and adhere to the tile 10. Screen 24 is then removed and paste 20 is allowed to dry as described below. A subsequent screen (not shown) also having locators (to enable registration of each print) but having different identification marks, in the form of different shapes,

dimensions, indicia or characters is then placed on tile 10. Fresh paste 20 (which may be different to the previously applied paste) is then urged through the second screen to form different labels on the tile or substrate 10. The process may then be repeated.

Tile 10 is then used to produce beads, for example, by laser cutting or "dicing" the tile 10. Tile 10 conveniently is sized such it comprises an array of 10,000 of the beads.

The paste may be rendered permanent by a firing, sintering or curing cycle or exposing it to radiation such as ultra violet radiation.

The paste 20 may comprise materials which may be screen printed: these include (organo-)metallic substances, cermets, ceramics, glasses and polymers. These materials show high stability under a range of chemical processes with little or no chemical interaction or leaching. Some pastes 20 have useful chemical activities such as acting as catalysts. For example, these include platinum or palladium bearing film.

Referring to Figures 2 to 8 there is now described an alternative method of making labelled articles. The alternative method may be combined with the previously mentioned method in order to provide a large number of articles of relatively large volume. Such articles may find specific application in certain types of chemical reactions. The method now described involves using photomasks. First screen 24 has an encoding of the form 103,123,---,---. The symbol "-" indicates that in the "digit" position screen 14 is arranged such that no paste 20 is printed. Screen 24 is termed the "high order screen". The high order screen 24 also has on its surface, (preferably in an area within

the flat tile 10 but outside of that area used for bead 12 formation), locators or a set of alignment patterns, which allow subsequent screens to be aligned in register with previously exposed patterns, images or indicia.

Tile or substrate 10 is then exposed in a first printing stage to transfer the pattern or indicia of the high order screen 24. The indicia are transferred as a set of printed paste features 20. The set of paste features 20 are then dried for example in an infra red tunnel (not shown). Optionally, tiles 10 may also be sintered or fired.

The next stage is to print or expose the tiles 10 using a so-called "low-order screen" 15. The low-order screen 15 bears encodings "---,---,--0,000" to "---,---,--9,999". The encoding patterns are arranged so that one low-order code section aligns against each bead position. Again the labels defined by paste 20 marks are dried and optionally fired or otherwise converted to their final state.

Two printing cycles of each of the 100 tiles 10 have been completed. All tiles 10 have been exposed to the same pattern. As will be apparent to those appropriately skilled, there is scope for considerable variation in the printing sequence described above. However, it is preferable that printing is undertaken as the first stage, as all tiles are identical at their completion. This facilitates the introduction of additional tiles.

Following the two printing cycles, each of the tiles 10 bears a set of encodings 003,123,--0,000 to 103,123,--9,999.

Either of two subsequent techniques may now be used. The first is for tiles 10 to be printed using a different screen bearing indicia so that each tile is distinguishable from example 100 screens bearing one of the encodings —,—,00,-,— to —,—,99-,—. Following this printing stage, paste 20 can be dried or fired to a final permanent form. Further printing stages may then be used.

Following this process tiles 10 are then re-divided into 10 lots in this case being for example: (0,10,20,30,40,50,60,70,80,90); (1,11,21,31,41,51,61,71,81,91) (9,19,29,39,49,59,69,79,89,99). Each of these lots of tiles 10 is then printed using a further screen (not shown) having an encoding of the form —,—,0--,— through —,—,9-- ,—. In this way each tile is exposed to a unique pairing of screens at the final exposure stage.

The end result is essentially identical to that previously described. However, the screen count required to obtain has been reduced from 102 to 22.

It is possible to use the above method with a single set of 10 screens, which are aligned with a controlled offset at each of two printing sequences. In doing this, the number of printing screens required is reduced to 12. However, greater precision of operation is required. Finally labelled beads or articles are formed by chopping or dicing the tile.

Variation may be made to the embodiments described, for example encoding of the form (0–9,A-Z) at each "digit" position may be used. This achieves 36 different indicia and a base 36 enumeration. In a presently preferred embodiment a set of symbols or indicia on the basis of their ready detection by a machine vision system may be used.

A low degree of common features between symbols is preferred, so that there is an inherent measure of redundancy in the symbols. Furthermore, copies of symbols or of error correction codes relating to groups of symbols (especially the high-order and low-order symbol groups) may be distributed within the area of the bead. In this way a robust system with a low read error rate and error checking facility is achievable .

An alternative method of manufacture will now be described with reference to figures 3 to 8, a bead 10 is defined; the bead 10 may be anything between approximately 250µm-10mm square, with a central region being approximately 800 µm square. The central region bears a unique code or signature. In practical embodiments this code most likely takes the form of a machine readable mark. This is typically a binary code or set of binary codes such as a two dimensional (2-D) bar code. However, for the sake of easy understanding, the code is assumed to take the form of conventional decimal digits in this description.

A set of 1,000,000,000,000 unique numbers (0-999,999,999,999) requires 12 digits for its representation as decimal numerals. It is assumed that substrate 12 used for bead

fabrication comprises a glass disc or silicon wafer of order 150 mm diameter. The disc or wafer is supported on an optical bench 13. Typically 10,000 beads may be made from such a disc or wafer Using conventional photolithographic techniques.

A first photomask 14, termed hereinafter the low order photomask, has a series of sequential numbers in the range 0000-9999 arranged in an array so that each number may be formed on each bead, for example in the lower, right hand corner of one face of the bead. Exposure of a substrate coated with a metalisation layer acts as a support on which visible code and a coat of a carefully selected photoresist 16 material is applied. For example, AZ5214E photoresist manufactured by Hoechst may be used for this purpose. Photoresist 16 is a positive working resist, capable of high resolution, in which the action of UV light from a source 18 generates radicals which are soluble in developer, but which cross-link to form an insoluble material on heating at around 120^0C. The property that this confers is that photoresist 16 may be "reversed" in its action from positive to negative working, locally in exposed areas, without any substantial deleterious effect on areas of unexposed photoresist 16. Heating to 120^0C for a short period is then performed. This is required to cross link (reverse) exposed areas of photoresist 16 and causes minimal degradation to the photosensitivity. Provided that the heating process is carried out within a reasonably short period after exposure, very high resolution is obtained.

Figure 8 shows diagrammatically a composite overlay of an active area of exposure fields for a bead to carry the code number 103123237514. This is formed from exposing the substrate to a high order photomask carrying the code 103123 in all positions, a low

order photomask carrying the code 7514 at the site corresponding to this bead, and two exposures to the middle order photomasks carrying in turn the codes 3 and 2 in all positions, aligned appropriately.

It will be appreciated that this process has used only a single coating of photoresist and only two develop cycles. A set of 11 base photomasks have been used, which are common to each lot of beads produced. A single photomask 14 is manufactured for each set of 1,000,000 beads. Variations to the present method are possible and will be apparent to those skilled in the art. For example, sets of 10,000,000 beads may be fabricated with a middle order photomask used in three locations with, 2500 beads being formed on each substrate. In order to do this a set of four low order photomasks are required.

The low order photomask is relatively difficult to manufacture, and therefore expensive, as each position has a different code. Its entire area is written by, for example, an electron beam. Other photomasks are identical in all positions, and thus the technique of step-and-repeat patterning can be utilised to facilitate their fabrication if this is desired. Therefore they are relatively easier to manufacture. A further advantage is that, provided the first photomask stage uses an exposure unit able to image substantially the whole of the substrate at any one time, then, the remaining stages are compatible with the use of a wafer stepper (reticle) exposure system.

It is apparent that by using a cell-based structure to build up the code, the requirement for alignment accuracy during photolithography is not overly onerous. This allows for

example, automatic alignment features and equipment to be used effectively, for example at the end of their useful life. Hence the method involves use of a relatively low-cost capital equipment.

It is not essential to form a complete encoding sequence. If process defects lead to unreadable encodings then this is of no consequence as such beads may be selectively discarded before chemical processing begins.

Below is described an alternative method of producing a set of beads bearing encodings represented by the numbers 103,123,000,000 to 103,123,999,999; that is a set of 1,000,000 encodings. The beads are to be produced initially in the form of a "large" flat substrate which is subsequently "diced" into individual beads. A relatively "large" flat substrate conveniently is sized so that it comprises an array of 10,000 of the beads.

As a first step all of the substrates are coated with a positive working photoresist, that is a material which is rendered selectively soluble in a developer medium by the local action of light, generally of UV light. A presently preferred resist is AZ5214E which is manufactured by the Hoechst company. Following resist coating all of the substrates are baked in the normal way.

All of the substrates are then exposed in a first exposure stage to transfer the pattern of the "low-order plate" as a latent image in the resist. Said low-order plate is designed so that it bears the encodings "---,---,--0,000" to "---,---,--9,999" arranged such that one low-order code section aligns to each of the bead positions. By way of example, Figure 2

shows a cell of the low order plate.All of the substrates are then exposed in a second exposure stage to a so-called "low-order plate". This image is then fixed by development, leaving resist in other areas of the encoded marks substantially unchanged. By way of example only, Figure 8 shows a cell of the high order plate. Once again the exposure process forms a latent image in the resist.

Again this latent image may be conveniently fixed by a process of development. Use may be made of a useful property of the AZ5214E resist which is presently preferred. Exposed areas of this resist may be "reversed" (i.e. rendered substantially insoluble to developer) and insensible to normal UV exposure levels by the simple expedient of a short baking cycle. This is achieved by placing the wafer on a hot plate maintained at 120°C for a period of order 120 seconds. The bake may marginally impair the sensitivity of the unexposed resist to subsequent UV exposure but not to a degree which impedes the process herein envisaged. It is noted that the baking process, if employed as is presently preferred, effectively reverses the action of the photoresist in response to UV light and that consideration should be given to this detail in the design of the photoplates.

Other materials may be used in "reversing" the action of resists, for example exposure to ammonia vapour. The term "reversing" it is intended to include other means as are well known to facilitate reversal.

The term image "fixing" includes development of the image or a reversal process or other such process as renders the image immutable. If all exposure cycles are completed rapidly there is no need to fix the image between the completion of the first exposure

cycle and final development. Fixing being employed primarily to avoid image degradation if substrates must be stored for protracted periods in a part exposed state.

Following the two exposure cycles we have 100 substrates each bearing a set of encodings 103,123,--0,000 to 103,123,--9,999.Optionally at this stage a cost benefit choice may be made. Either:

1) Each substrate may be exposed to a separate mask, using 100 masks bearing the encodings ---,---,00,-,--- to ---,---,99-,---. Following this stage, given care in the detail of the mask design, the resist is developed and this yields the required set of 1,000,000 unique pattern. In this way a total of 102 masks have been used. However, to form a next or subsequent set of differently encoded beads only the single "high-order mask" needs to be replaced;

2) Two further exposure stages may be used. In the first exposure stage the substrates are divided into 10 lots, for example 0–9, 10–19, 20–29...90–99. Each lot of substrates is exposed to a mask bearing a code of the form ---,---,-0-,--- through ---,---,-,9-,---. By way of example substrates 0–9 are exposed to ---,---,-0-,---, 10–19 to ---,---,-1-,--- and so on up to and including exposing 90–99 to ---,---,-9-,---.

Following this process the substrates are then re-divided into 10 lots in this case being for example:(0,10,20,30,40,50,60,70,80,90);(1,11,21,31,41,51,61,71,81,91)...(9,19,29,39,49 ,59,69,79,89,99).Each of these lots of substrates is then exposed to a further mask having an encoding of the form ---,---,0--,--- through ---,---,9--,---. In this way each substrate is exposed to a unique pairing of masks. By way of example only Figures 8, 9 show cells of

these "middle order" masks.The end result is essentially identical to that previously described, however, the mask count has been reduced from 102 to 22. Again maintaining the advantage that only a single plate has to be fabricated to produce a next or subsequent set of unique beads. By way of example only , a composite image resulting from the process is shown in Figure 8.

It is also possible to use the method above with a single set of 10 masks, which are aligned with a controlled offset at each of the two exposure sequences. In so doing, the number of masks required is further reduced to 12.

Whichever scheme is employed the final stage is to flood expose the resist and then develop the image. Dicing of the beads may be achieved by laser cutting or chemical etching.

Alternate numbering schemes may be employed. For example encoding of the form (0–9,A-Z) at each "digit" position may be used. In a presently preferred embodiment a set of symbols can be selected on the basis of their ready detection by a machine readable system, preferably so designed as to have a low degree of common features between the symbols such that there is an inherent measure of redundancy. Furthermore, error correction codes relating to groups of symbols (especially the high-order and low-order symbol groups) are distributed within the area of the bead. In this way the method has a low read error rate in application.

It is understood that the above two methods of manufacturing uniquely labelled articles can be combined.

CLAIMS

1. A method of fabricating a plurality of articles, each having a unique label, comprising the steps of: defining the articles on a substrate; forming or causing to be formed on articles, of at least two substrates a first indicia sufficient to identify, on each substrate, articles one from another; forming or causing to be formed, a second indicia on articles on one of said substrates sufficient to identify articles of one substrate from articles of another substrate; and disassembling the substrates into a plurality of articles.

2. A method according to claim 1 wherein the articles are formed in an array and the first indicia are formed on the array.

3. A method according to claim 2 wherein the second indicia formed on each article of a substrate are all identical.

4. A method according to any preceding claim wherein third or subsequent indicia are formed on each article.

5. A method according to any preceding claim wherein at least the first indicia are applied to a substrate by screen printing.

6. A method according to any of claims 1 to 4 wherein at least first indicia are applied to a substrate by exposing the substrate to radiation through a photomask.

7. A method according to claim 6 wherein indicia are transfered to one or more underlying layer(s) by etching.

8. A method according to any of claims 5 to 7 wherein second indicia are applied to a substrate by exposing the substrate to radiation through a photomask.

9. A method according to claim 5 wherein first indicia are defined by a paste.

10. A method according to claim 9 wherein the paste is dried, heated, fired or cured.

11. A method according to any preceding claim wherein first and second indicia are located at different locations on each article.

12. A method according to any preceding claim wherein indicia are located at different orientations.

13. A method according to any preceding claim wherein articles are formed in an array comprising 10 - 100 rows and 10 – 100 columns.

14. A method according to claim 6 wherein registration of subsequent photomasks is achieved by aligning features on the photomask and substrate.

15. A method of manufacturing a plurality of uniquely labelled articles comprising the steps of: applying a medium, via a first printing screen to a tile; removing the first printing screen presenting the tile to a second printing screen and applying a medium via said second printing screen to said tile; curing said tile and cutting the tile into a plurality of uniquely labelled articles.

16. A method of manufacturing a plurality of uniquely labelled articles, comprising the steps of: exposing a photoresist coated substrate, from which articles are to be formed, using a first photomask to define a first set of unique low order indicia at a plurality of regions at first locations on the substrate; subsequently exposing the substrate to a second photomask to define a high order indicia at second locations on the substrate so that combinations of low order indicia and high order indicia are unique for each article and disassembling the substrate into said plurality of labelled articles.

17. A plurality of uniquely labelled articles fabricated according to any of claims 1 to 15.

18. A method substantially as herein described with reference to the Figures.

19. Apparatus for manufacturing a plurality of uniquelly labelled articles comprises means for forming a plurality of articles on a substrate, means for labelling each article with a first indicia, means for labelling each article with a second indicia and means for dissassembling the labelled articles.

20. Articles substantially as herein described with reference to the Figures.

Fig.1.

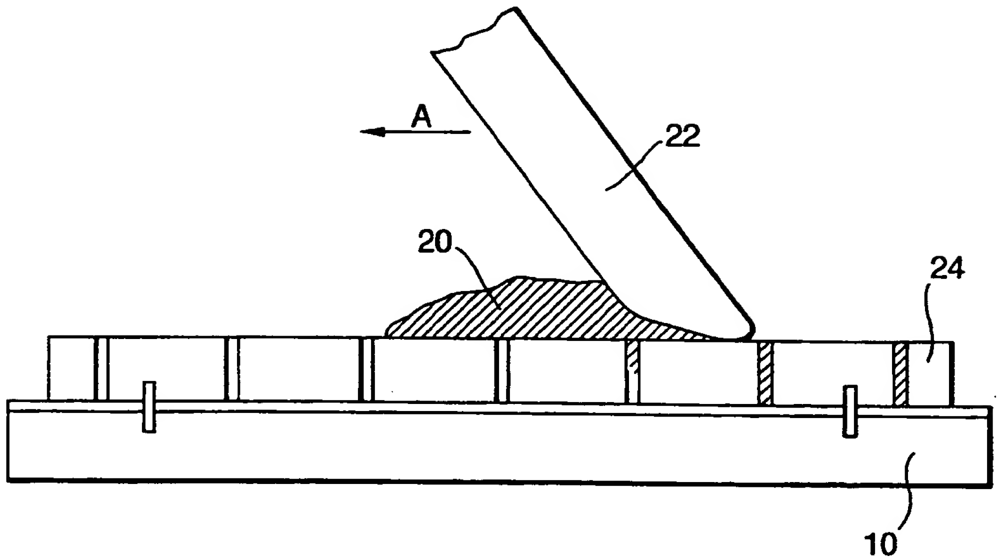


Fig.2a.

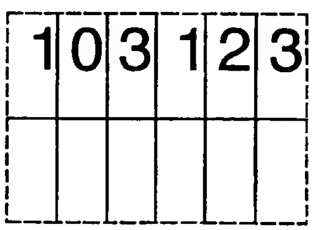


Fig.2b.

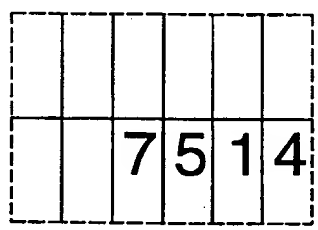


Fig.2c.

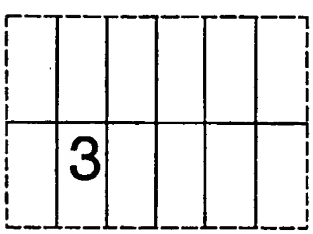


Fig.2d.

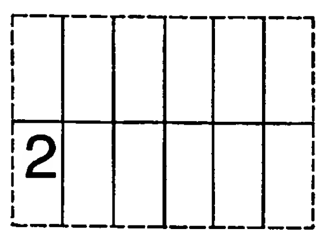


Fig.3.

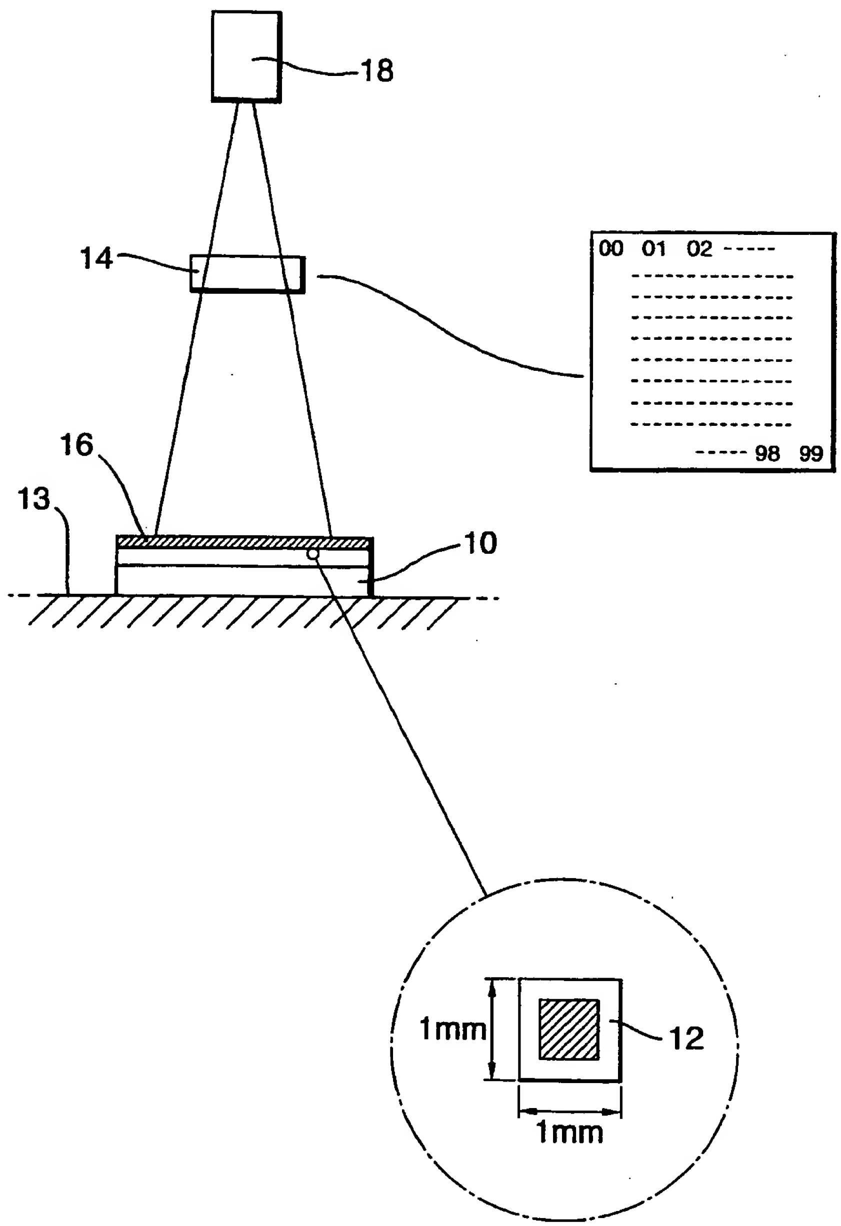


Fig.4.

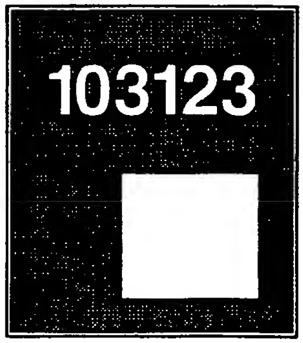


Fig.5.

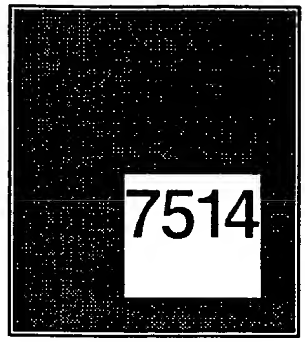


Fig.6.

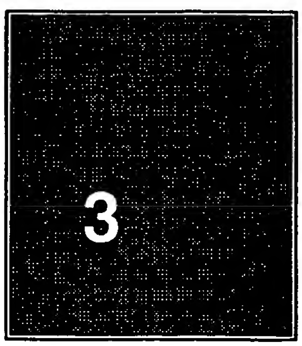


Fig.7.

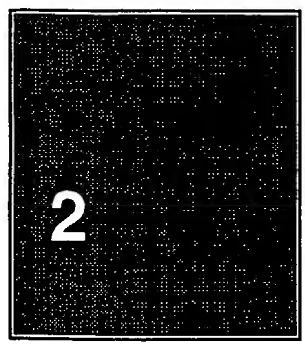
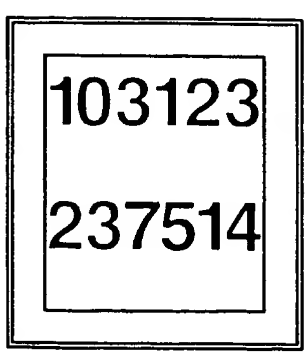


Fig.8.



INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 99/00762

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 B01J19/00 B01J13/02 G03F7/00 G09F3/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 B01J G06K G09F G03F B81C B81B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 4 613 230 A (HIROSHI IWAI) 23 September 1986	1-3,6,8, 10,11, 14,16, 17,19
	see column 7, line 22 - column 9, line 37 see figure 11	
A		7,12,13, 15

A	US 4 343 877 A (PING-WANG CHIANG) 10 August 1982 see column 8, line 13 - column 10, line 57 see figures ---	1-4,6-8, 10-17,19
A	US 5 314 829 A (L. STEPHEN COLES & MARINA DEL REY) 24 May 1994 see the whole document ---	1,2,6-8, 11,13, 16,17,19

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
18 May 1999	04/06/1999

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Stevnsborg, N

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INTERNATIONAL SEARCH REPORT

Internal Application No
PCT/GB 99/00762

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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A	PATENT ABSTRACTS OF JAPAN vol. 13, no. 578 (E-864), 20 December 1989 & JP 01 241815 A (SEIKO EPSON CORP.), 26 September 1989 see abstract; figure	1,2,6-8,13,16,17
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A	P.H. KAYE ET AL.: "The Production of Precision Silicon Micromachined Non-spherical Particles for Aerosol Studies" JOURNAL OF AEROSOL SCIENCE, vol. 23, no. Suppl. 1, 1992, pages s201-s204, XP002102951 Oxford, GB see the whole document	

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